(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 0 736 226 B1

(12) EUROPEAN PATENT SPECIFICATION

(45) Date of publication and mention of the grant of the patent:
24.03.1999 Bulletin 1999/12

(21) Application number: 95902722.8

(22) Date of filing: 07.12.1994

(51) Int Cl.[6]: H01M 8/04, H01M 8/24

(86) International application number:
PCT/CA94/00675

(87) International publication number:
WO 95/17772 (29.06.1995 Gazette 1995/27)

(54) ELECTROCHEMICAL FUEL CELL EMPLOYING AMBIENT AIR AS THE OXIDANT AND COOLANT

ELEKTROCHEMISCHE BRENNSTOFFZELLE MIT VERWENDUNG VON LUFT ALS OXIDANT UND KÜHLUNG

CELLULE ELECTROCHIMIQUE DANS LAQUELLE L'AIR AMBIANT EST UTILISE COMME OXYDANT ET REFRIGERANT

(84) Designated Contracting States:
CH DE FR GB IT LI SE

(30) Priority: 22.12.1993 US 171732

(43) Date of publication of application:
09.10.1996 Bulletin 1996/41

(73) Proprietor: BALLARD POWER SYSTEMS INC.
Burnaby, British Columbia V5J 5J9 (CA)

(72) Inventors:
- FLETCHER, Nicholas J.
  Vancouver, British Columbia V6R 1A5 (CA)
- LAMONT, Gordon J.
  Vancouver, British Columbia V5Y 2J5 (CA)
- BASURA, Vesna
  Burnaby, British Columbia V5G 4K5 (CA)
- VOSS, Henry H.
  West Vancouver, British Columbia V7T 1H1 (CA)
- WILKINSON, David P.
  North Vancouver, British Columbia V7R 1W (CA)

(74) Representative: Haecker, Walter, Dipl.-Phys. et al
Hoeger, Stellrecht & Partner
Uhlandstrasse 14 c
70182 Stuttgart (DE)

(56) References cited:
DE-C- 2 140 988
US-A- 3 623 913
US-A- 3 743 544

- PATENT ABSTRACTS OF JAPAN, vol. 11, no. 113(E-497); & JP,A,61260551 (FUJI ELECTRIC CO LTD), 18-11-1986
- PATENT ABSTRACTS OF JAPAN, vol. 12, no. 320(E-651); & JP,A,6386270 (HITACHI LTD), 16-04-1988

EP 0 736 226 B1

Printed by Jouve, 75001 PARIS (FR)

Description

Field Of The Invention

[0001] This invention relates generally to electrochemical fuel cells and, more particularly, to a fuel cell which employs ambient air as both an oxidant and a coolant.

Background Of The Invention

[0002] A fuel cell is a device which generates electrical energy by converting chemical energy directly into electrical energy by oxidation of fuel supplied to the cell. Fuel cells are advantageous because they convert chemical energy directly to electrical energy without the necessity of undergoing any intermediate steps, for example, combustion of a hydrocarbon or carbonaceous fuel as takes place in a thermal power station.

[0003] A typical fuel cell includes an anode, a cathode and an electrolyte. Fuel and oxidant are supplied to the anode and cathode, respectively. At the anode, the fuel permeates the electrode material and reacts with an anode catalyst layer to form cations (protons) and electrons. The cations migrate through the electrolyte to the cathode. At the cathode, the oxygen-containing gas supply reacts with a cathode catalyst layer to form anions. The electrons produced at the anode travel from the fuel cell anode, through an external load, and back into the cathode of the cell. The anions produced at the cathode react with the cations and electrons to form a reaction product which is removed from the cell.

[0004] In electrochemical fuel cells employing hydrogen as the fuel and oxygen-containing air (or pure oxygen) as the oxidant, a catalyzed reaction at the anode produces hydrogen cations from the fuel supply. This type of fuel cell is advantageous because the only reaction product is water. An ion exchange membrane facilitates the migration of hydrogen cations from the anode to the cathode. In addition to conducting hydrogen cations, the membrane isolates the hydrogen fuel stream from the oxidant stream comprising oxygen containing air. At the cathode, oxygen reacts at the catalyst layer to form anions. The anions formed at the cathode react with the hydrogen ions that have crossed the membrane to form liquid water as the reaction product. The anode and cathode reactions in such fuel cells is shown in the following equations:

Anode reaction: $H_2 \rightarrow 2H^+ + 2e^-$

Cathode reaction: $1/2O_2 + 2H^+ + 2e^- \rightarrow H_2O$

[0005] A type of fuel cell known as a solid polymer fuel cell ("SPFC") contains a membrane electrode assembly ("MEA") consisting of a solid polymer electrolyte or ion exchange membrane disposed between two electrodes formed of porous, electrically conductive sheet material. The electrodes are typically formed of carbon fiber paper ("CFP"), and are generally impregnated or coated with a hydrophobic polymer, such as polytetrafluoroethylene. The MEA contains a layer of catalyst at each membrane/electrode interface to induce the desired electrochemical reaction. A finely divided platinum catalyst is typically employed. The MEA is in turn disposed between two electrically conductive plates, each of which has at least one flow passage engraved or milled therein. These fluid flow field plates are typically formed of graphite. The flow passages direct the fuel and oxidant to the respective electrodes, namely, the anode on the fuel side and the cathode on the oxidant side. The electrodes are electrically coupled to provide a path for conducting electrons between the electrodes.

[0006] In a single cell arrangement, fluid flow field plates are provided on each of the anode and cathode sides. The plates act as current collectors, provide support for the electrodes, provide access channels for the fuel and oxidant to the respective anode and cathode surfaces, and provide channels for the removal of water formed during operation of the cell.

[0007] Two or more fuel cells can be connected together in series or in parallel to increase the overall power output of the assembly. In such arrangements, the cells are typically connected in series, wherein one side of a given plate serves as an anode plate for one cell and the other side of the plate is the cathode plate for the adjacent cell. Such a series connected multiple fuel cell arrangement is referred to as a fuel cell stack, and is usually held together by tie rods and end plates. The stack typically includes manifolds and inlets for directing the fuel (substantially pure hydrogen, methanol reformate or natural gas reformate) and the oxidant (substantially pure oxygen or oxygen-containing air) to the anode and cathode flow field channels. The stack also usually includes a manifold and inlet for directing the coolant fluid, typically water, to interior channels within the stack to absorb heat generated by the exothermic reaction of hydrogen and oxygen within the fuel cells. The stack also generally includes exhaust outlets and manifolds for expelling the unreacted fuel and oxidant gases, each carrying entrained water, as well as an outlet manifold for the coolant water exiting the stack.

[0008] Conventional fuel cell and stack designs have several inherent disadvantages. First, conventional designs typically employ liquid cooling systems for regulating the cells' operating temperature. Liquid cooling systems are disadvantageous because they require the incorporation of additional components to direct coolant into thermal contact with fuel cells. The power requirements to operate such additional components, such as pumps and cooling fans, represent an additional parasitic load on the system, thereby decreasing the net power derivable from the stack. Such additional components also add volume, weight, complexity and cost to

fuel cell designs.

[0009] Second, conventional designs employ further parasitic devices such as pumps for the delivery of pressurized fuel and oxidant to the fuel cell. In addition to adding volume, weight, complexity and cost, these parasitic systems also reduce the overall power efficiency of the system.

[0010] Third, in conventional stack arrangements it is difficult to identify and replace defective fuel cells without disrupting the operation of the entire fuel cell stack.

[0011] Because the production of electrical energy in a fuel cell is an exothermic reaction, such fuel cell needs to be cooled in order to be kept at a desired operating temperature, and as mentioned before, preferably ambient air is used as a coolant.

[0012] DE-A-21 40 988 discloses a stack of fuel cells; each fuel cell has an electrode-electrolyte assembly comprising a porous plate-shaped anode, a porous plate-shaped cathode and a plate-shaped electrolyte member interposed between the anode and the cathode, said electrolyte member having a porous PTFE plate and an acid retained within the pores of the PTFE plate. Each of said electrode-electrolyte assemblies has an impervious plate for current collection, reactant distribution and temperature control of the fuel cell. In order to achieve these functions said impervious plate is made from a metal, has a first major surface provided with a series of first laterally spaced ribs abutting against the cathode of the fuel cell, and a second major surface provided with a series of second laterally spaced ribs abutting against the anode of the next fuel cell. The first ribs form air flow channels between the cathode of the fuel cell and said impervious plate, and the second ribs extending perpendicularly to the first ribs form fuel flow channels between said impervious plate and the anode of the next fuel cell. Ambient air is flown through said air flow channels in order to serve as an oxidant and as a coolant so that the first ribs contacting the cathode of the fuel cell also form cooling fins for dissipating heat generated in the fuel cell.

[0013] Because at least most of the fuel introduced between the second ribs into the fuel flow channels is consumed for producing electrical energy, the impervious plate is not effectively cooled at its side facing away from the cathode.

[0014] US-A-3 623 913 discloses a stack of fuel cells which differ from the fuel cells described in DE-A-21 40 988 only in that (i) said impervious metal plate of each fuel cell has a fin-like extension projecting beyond one side of the fuel cell stack, and (ii) the side of said impervious plate facing the cathode has a crisscross pattern of grooves formed by the air flow channels and by a series of laterally spaced grooves extending perpendicularly to said air flow channels and over said fin-like extension.

[0015] JP-A-63-86270 discloses fuel cells with cooling fins provided at one of the two major surfaces of a plate and being in contact with the atmosphere, and JP-A-61-260551 discloses fuel cells with cooling fins provided at projections extending into an air duct disposed at one side of a fuel cell stack and being provided with a fan for conveying ambient air through said duct.

[0016] It is an object of the present invention to propose an electrochemical fuel cell assembly employing ambient air as an oxidant and coolant, in which heat generated by the exothermic reaction for producing electrical energy is more effectively dissipated to the atmosphere.

[0017] This object is achieved by the fuel cell assembly of claim 1.

[0018] In operation, heat generated in the fuel cell assembly is effectively dissipated to the atmosphere at both sides of the thermally conductive plate.

[0019] The thermally conductive plate is preferably, but not necessarily, formed as a single planar piece from which the thermally conductive members extend. Alternatively, the plate could consist of a plurality of staggered bars interconnecting the thermally conductive members, which extend from the staggered bars and contact the exposed cathode surface.

[0020] The plate and the first members are preferably formed of aluminum, and the portions of the first members which contact the cathode surface have an inert metal applied thereto. The inert metal is preferably gold applied by electroplating.

[0021] The preferred electrical connection means comprises electrical conductors disposed between the anode and the ion exchange membrane, and the electrical conductors preferably extend through the sealing means. The preferred electrical conductors are formed from gold wire.

[0022] In the preferred assembly, the thermally conductive material extending from the second major surface of the plate comprises a plurality of thermally conductive members, or alternatively a thermally conductive foam. The preferred thermally conductive foam is an aluminum foam.

[0023] In the preferred assembly, the fuel delivery means comprises a fuel inlet and a fuel outlet, such that the fuel outlet directs unreacted components of the gaseous fuel stream away from the anode. The assembly can further comprise a fan for directing the ambient air onto the exposed surface of the porous electrically conductive cathode. Where the gaseous fuel stream comprises hydrogen, the assembly preferably further comprises means for accumulating water condensed on the first thermally conductive members.

[0024] The present invention also relates to a fuel cell stack comprising a plurality of the above fuel cell assemblies and having the features of claim 13.

[0025] The fuel cell stack can be formed as a multiplexed arrangement, wherein the plurality of fuel cell assemblies share a common ion exchange membrane.

[0026] According to the present invention, the above object can be also achieved by an electrochemical fuel cell assembly comprising a bicell membrane electrode

assembly and having the features of claim 15.

[0027] In operation, at least a portion of the heat generated exothermically in the bicell membrane electrode assembly is dissipated to the atmosphere through the first and second members.

[0028] Such bicell membrane electrode assembly may have the features of one or several of claims 2 to 14.

Brief Description of The Drawings

[0029] **FIG. 1** is an exploded perspective view of an electrochemical fuel cell assembly employing ambient air as the oxidant and coolant.

[0030] **FIG. 2A** is a section view taken in the direction of arrows 2-2 in **FIG. 1**.

[0031] **FIG. 2B** is section view of an alternative embodiment of the electrochemical fuel cell assembly illustrated in **FIGS. 1 and 2A.**

[0032] **FIG. 3** is a perspective view of a fuel cell stack connected across an external load.

[0033] **FIGS. 4A and 4B** illustrate alternative embodiments of an interleaved membrane electrode assembly according to the present invention.

[0034] **FIG. 5** is a side sectional view of an alternative embodiment of an electrochemical fuel cell assembly employing ambient air as the oxidant and coolant.

[0035] **FIG. 6** is a sectional view of a multiplexed arrangement of three bicell membrane electrode assemblies employing ambient air as the oxidant and coolant, which share common ion exchange membranes.

[0036] **FIG. 7** is an exploded perspective view of a first embodiment of a thermally conductive member or fin subassembly for an electrochemical fuel cell assembly employing ambient air as the oxidant and coolant, which employs a slidable comb for adjusting the configuration of the air conducting channels.

[0037] **FIG. 8** is a perspective view of second embodiment of a thermally conductive member or fin subassembly for an electrochemical fuel cell assembly employing ambient air as the oxidant and coolant, which employs a pivotable baffle (shown in phantom lines) for adjusting the flow through the air conducting channels.

[0038] **FIG. 9** is a perspective view of a pivotable baffle subassembly for use in conjunction with the fin subassembly shown in **FIG. 8**.

[0039] **FIG. 10** is a side view of the pivotable baffle subassembly shown in **FIG. 9.**

[0040] **FIG. 11** is a schematic view of third embodiment of an electrochemical fuel cell assembly employing ambient air as the oxidant and coolant, which employs external dampers for adjusting the flow through the air conducting channels.

Detailed Description Of The Preferred Embodiments

[0041] Referring first to **FIG. 1** and **FIG. 2A**, an electrochemical fuel cell assembly **10**, includes a bicell membrane electrode assembly ("MEA") **14.** Bicell MEA **14** includes a first cathode **16**, an anode **26**, and a second cathode **38.** A first ion exchange membrane **24** is interposed between first cathode **16** and anode **26.** A second ion exchange membrane **34** is interposed between second cathode **38** and anode **26**. Fuel supply line **44** and fuel inlet **46** contain and direct fuel at a pressure slightly greater than atmospheric to anode **26.**

[0042] The electrodes **16, 26, 38** are formed of porous electrically conductive sheet material, preferably porous carbon fiber paper ("CFP") impregnated or coated with a hydrophobic polymer, such as polytetrafluoroethylene. The electrodes **16, 26, 38** are each treated with a layer of catalyst, such as platinum or other suitable electrocatalytic material, on the surface(s) adjacent and in contact with the ion exchange membrane(s) **24, 34** to facilitate the desired chemical reaction. Suitable ion exchange membranes are commercially available from DuPont under the trade name Nafion 117 and from Dow under the trade designation XUS 13204.10.

[0043] The electrodes **16, 26, 38** and the ion exchange membranes **24, 34** are arranged together in an interleaved or sandwich-like manner, as illustrated in **FIG. 1** and **FIG. 2A**, and placed in a high pressure press at a temperature sufficient to soften the ion exchange membrane material. The combination of pressure and temperature forces the softened membrane material at least partially into the CFP electrode material, bonding the individual layers to form a single unitary assembly. Presently, the bicell MEA **14** is formed by placing the layers of material in a press at a temperature and pressure sufficient to soften the material and create an intimate bond.

[0044] Low pressure can be employed to supply the gaseous fuel because the chemical reaction at the anode **26** consumes the fuel and draws it into the anode **26.** The porous structure of the CFP used to form the anode **26** facilitates the delivery of the gaseous fuel throughout the anode **26.** The gaseous fuel reacts at the anode **26** to produce cations (protons) and electrons. When hydrogen is used as the fuel, the reaction at the anode produces hydrogen cations and electrons according to the following equation:

$$H_2 \rightarrow 2H^+ + 2e^-.$$

[0045] The reaction at the cathodes **16, 38** produces water according to the following equation:

$$1/2\ O_2 + 2H^+ + 2e^- \rightarrow H_2O.$$

The ion exchange membrane facilitates the migration of cations from the anode **26** to the cathodes **16, 38.** In addition to conducting hydrogen cations, the ion exchange membranes **24, 34** isolate the gaseous fuel stream from the oxidant stream. This is particularly im-

portant when hydrogen is employed as a fuel source because of the reaction which occurs when hydrogen and oxygen are mixed and ignited or contacted with a catalyst.

[0046] A seal 50 provides a gas-impermeable barrier at the edges of the anode **26** to prevent leakage of the gaseous fuel from within anode **26.** In **FIG. 2A,** the seal **50** is formed by extending the ion exchange membranes **24, 34** over the edges of the anode **26**. During the assembly process, the portions of the ion exchange membranes **24, 34** extending over the anode **26** can be adhered using heat and pressure to form a gas-impermeable seal around the anode **26.** Alternatively, as illustrated in **FIG. 2B**, the seal 50 may be formed by disposing layers of sealant **52a, 52b**, such as a silicon based sealant, along the top and bottom edge portions, respectively, of anode **26** which extend between the ion exchange membranes **24, 34.**

[0047] As shown in **FIGS. 1, 2A** and **2B**, edge current collectors **56a, 56b** are disposed between the anode **26** and the ion exchange membranes **24, 34**. The first edge current collector **56a** is disposed between the anode **26** and the first ion exchange membrane **24**, and the second edge current collector **56b** is disposed between the anode **26** and the second ion exchange membrane **34.** The edge current collectors **56a, 56b** facilitate current flow (i.e., electron flow) from the anode **26** to an external load, as described in more detail below. As best shown in **FIG. 2A**, the edge current collectors **56a, 56b** exit the bicell MEA **14** through the seal **50,** thereby providing an electrical connection to the anode **26.**

[0048] Each of the edge current collectors **56a, 56b** is preferably formed from a plurality of electrically conductive wires (not shown). The wires forming the edge current collectors **56a, 56b** are in turn preferably formed from a highly conductive material such as gold, niobium, platinum, titanium or graphite. Although a single wire can provide sufficient edge current collection, a plurality of wires is preferred. In **FIG. 1**, the conductive wires **56a, 56b** are shown exiting from both the top and bottom of the bicell MEA **14**, whereas in **FIGS. 2A** and **2B** the conductive wires only exit from the top of the bicell MEA **14.**

[0049] As shown in **FIGS. 1, 2A** and **2B**, the fuel cell assembly **10** further includes first and second thermally conductive plates **62a, 62b** disposed on opposite sides of the bicell MEA **14**. The plates **62a, 62b** are preferably constructed from aluminum which is either milled or extruded to form the illustrated configuration. Aluminum is preferred because it is relatively inexpensive and lightweight and because it has favorable thermal and electrical conductivity.

[0050] As shown in **FIG. 1**, each plate **62a, 62b** includes a first set of thermally conductive members, shown in **FIG. 1** as fins **66a, 66b**, which extend toward the bicell MEA **14** and contact one of cathodes (cathode **38** in **FIG. 1** and **FIG. 2A**) and a second set of thermally conductive members, shown in **FIG. 1** as fins **64a, 64b**, which extend away from the bicell MEA **14**. The portion of each fin **66a, 66b** which contacts the surface of a cathode is preferably plated with gold to prevent oxidization of the aluminum and ensure good electrical contact between the cathode **38** and each fin **66a, 66b.**

[0051] The first set of thermally conductive members **66a, 66b** provide structural rigidity and support for the bicell MEA **14**, stabilize the MEA **14**, and inhibit distortion of the MEA **14** from swelling due to oversaturation of the membrane.

[0052] Each of the second set of thermally conductive members, shown in **FIG. 1** as fins **64a, 64b**, could also be formed as a thermally conductive foam, in lieu of the fins. Thermally conductive foam has an irregular three-dimensional conformation, with interstitial spaces permitting the passage of air and other coolant fluids through the irregular, lattice-like structure of the thermally conductive material from which the foam if formed. The preferred thermally conductive foam is an aluminum foam.

[0053] As shown in **FIG. 1**, a fastener mechanism secures the plates **62a, 62b** and MEA **14** in assembled form and maintains contact between the fins **66a, 66b** and the exposed surfaces of cathodes **16, 38.** The fastener mechanism preferably includes a first threaded fastener **72** extending through the upper portion of the plates **62a, 62b** and a second threaded fastener **74** extending between the bottom portion of the plates **62a, 62b.** The threaded fasteners **72, 74** connect the plates **62a, 62b** and allow the plates **62a, 62b** to be clamped against the bicell MEA **14**, thereby maintaining electrical and physical contact between the cathodes **16, 38** and the plates **62a, 62b.**

[0054] Both sets of fins **64a, 64b** and **66a, 66b** are open at the top and bottom to allow air flow through the fins. Heat produced by the exothermic chemical reaction of fuel (hydrogen) and oxidant (oxygen) within the bicell MEA **14** is dissipated to the atmosphere through the fins **64a, 64b** and **66a, 66b.** It has been found that such heat dissipation produces a natural convection current which causes the ambient air to be drawn upwardly through the fins **64a, 64b** and **66a, 66b**. The set of fins **64a** extend in a direction away from MEA **14**, and function primarily as heat transfer surfaces for expelling waste heat to the atmosphere such that a desired operation temperature of the bicell MEA **14** is maintained. The sets of fins **66a, 66b**, in addition to functioning as heat transfer surfaces, cooperate with the plates **62a, 62b** and the adjacent cathodes to form a plurality of air conducting channels which draw oxygen-containing ambient air toward the exposed surface of the cathodes. For example, fins **66a** cooperate with plate **62a** and cathode **16** to form an air conducting channel **78** (see **FIG. 1**). A similar plurality of air conducting channels draws oxygen-containing ambient air toward the exposed surface of cathode **38.** The vertical orientation of the air supply channels **78** allows the water produced at the cathode **16** to flow downwardly toward the bottom of the fuel cell assembly **10** where it can be drained from the assembly, thereby

preventing oversaturation of the ion exchange membrane **24**.

[0055] In employing ambient air as the oxidant and coolant for the fuel cell assembly **10**, the following operating conditions should be present:

(1) ambient air flow through the air conducting channels to provide a sufficient stoichiometric supply of reactant oxygen to support the reaction at the membrane electrode assembly;
(2) ambient air flow and operating temperature should be such that the water removal capacity of the ambient air flow is less than the rate of production of reactant water to prevent dehydration of the ion exchange membranes;
(3) the operating temperature of the cell should be high enough to provide reasonable fuel cell performance; and
(4) the operating temperature of the fuel cell should be high enough to allow the cell to reject waste heat to the atmosphere by natural convection.

[0056] With these considerations in mind, the size, spacing, and number of members or fins is empirically optimized to provide temperature stability and performance stability over a wide range of loads.

[0057] Turning now to **FIG. 3**, a plurality of the fuel cell assemblies, six of which are designated in **FIG. 3** as assemblies **10a, 10b, 10c, 10d, 10e,** and **10f**, can be combined to form a fuel cell stack **100**. Fuel inlets, one of which is designated in **FIG. 3** as fuel inlet **146**, each direct a fuel stream to one of the respective fuel cell assemblies **10a-f**. The fuel inlets are connected to a main fuel supply line **104**, which is in turn connected to a fuel supply source (not shown) for delivering gaseous fuel at a pressure slightly greater than atmospheric to the stack **100**.

[0058] In **FIG. 3**, the fuel cells assemblies **10a-f** are electrically connected in series so that the fuel cell stack **100** produces a voltage potential equal to the sum of the voltages of the individual fuel cell assemblies **10a-f**. More specifically, the edge current collectors **156** are used to electrically couple the anode of one bicell MEA to the cathodes of the next adjacent bicell MEA in the stack **100**. For example, in **FIG. 3** the anode of the first fuel cell assembly **10a** is electrically connected to the cathodes of the second fuel cell assembly **10b**. This electrical connection is preferably made by connecting the edge current collectors **156** from one fuel cell assembly to the plate **162** adjacent the next fuel cell assembly in the stack **100**.

[0059] The full electrical potential of the stack **100** is imposed between a positive lead **108** and a negative lead **110**. The positive lead **108** is formed by connecting an electrical conductor **112** to a positively charged portion of the first cell **10a** in the stack **100**. Specifically, the positive lead **108** can be connected to either of the end plates, the fins, the threaded fasteners, or the cathodes of the first cell **10a**. The negative lead **110** is formed by joining the edge current collectors of the last fuel cell assembly **10f** to form a single conductor **114**.

[0060] As is illustrated schematically in **FIG. 3**, when the stack **100** is installed in an electrical circuit, a load **118** and a contactor switch **120** can be connected between the positive and negative leads **108, 110**. The contactor switch **120** can be selectively opened and closed to deliver power from the stack **100** to the load **118**.

[0061] **FIGS. 4A** and **4B** illustrate alternative embodiments for serially connecting individual bicell MEAs to form a stack configuration. In both **FIGS. 4A** and **4B**, the electrodes of successive bicell MEAs are interleaved to form serial electrical connections. Each bicell MEA **114** includes a center anode **116** interposed between two cathodes **120, 122**. Two sheets of solid polymer ion exchange membranes **126, 128** are interposed between the anode **116** and the cathodes **120, 122**. In **FIG. 4A**, sealant material **132** is disposed at both ends of the anode **116** to prevent leakage of the gaseous fuel supplied to the anode **116**. In **FIG. 4B**, a single sheet of material is used to form ion exchange membranes **126, 128**. The membrane material is looped around one end of the anode **116** and sealant material **132** is used to seal the other end of the anode **116**.

[0062] In both embodiments illustrated in **FIGS. 4A** and **4B**, the cathodes **120, 122** extend beyond one end of a respective anode **116** and are joined around an electrical conductor **136**. The electrical conductor **136** in turn extends through the sealant **132** and into the anode **116** of the next bicell MEA **114b** in the stack.

[0063] **FIG. 5** illustrates an alternative embodiment of a fuel cell assembly which employs ambient air as the oxidant and coolant. In **FIG. 5**, a unicell MEA **214** is employed as opposed to the bicell MEA arrangement of FIGS. **1, 2A** and **2B**. MEA **214** includes an ion exchange membrane **224**, which is interposed between anode **226** and cathode **216**. A seal **250**, formed of sealant material disposed along the exterior surfaces of the anode **226**, is also shown in **FIG. 5**. Seal **250** forms a gas-impermeable barrier to prevent leakage of gaseous fuel supplied to the anode **226**. A fuel delivery mechanism **244** delivers gaseous fuel (preferably substantially pure hydrogen) to the anode **226** of the unicell MEA **214**. The fuel delivery means **244** includes at least one fuel inlet **246** which extends partially into the anode **226**. The fuel inlet **246** delivers gaseous fuel to the anode **226** at a low pressure or at slightly greater than atmospheric pressure.

[0064] In the embodiment illustrated in **FIG. 5**, a clamping mechanism **218** secures the plate **262**, together with its fins **264, 266**, against the cathode **216** of the unicell MEA **214**. The clamping means **218** is illustrated in **FIG. 5** as a pair of threaded fasteners **272, 274** and an end plate **220**.

[0065] **FIG. 6** shows a multiplexed arrangement **302** of three bicell assemblies employing ambient air as the

oxidant and coolant. The multiplexed arrangement includes first cathodes **304a, 304b, 304c**, anodes **306a, 306b, 306c**, and second cathodes **314a, 314b, 314c**. As shown in **FIG. 6**, first cathode **304a**, anode **306a** and second cathode **314a** are arranged in a first bicell MEA **310a**, with first ion exchange membrane **316** interposed between first anode **306a** and cathode **304a**, and second ion exchange membrane **326** interposed between anode **306a** and second cathode **314a**. Similarly, first cathode **304b**, anode **306b** and second cathode **314b** are arranged in a second bicell MEA **310b**, with first ion exchange membrane **316** interposed between first anode **306b** and cathode **304b**, and second ion exchange membrane **326** interposed between anode **306b** and second cathode **314b**. Finally, first cathode **304c**, anode **306c** and second cathode **314c** are arranged in a third bicell MEA **310c**, with first ion exchange membrane **316** interposed between first anode **306c** and cathode **304c**, and second ion exchange membrane **326** interposed between anode **306c** and second cathode **314c**. As shown in **FIG. 6**, first, second and third bicell assemblies **310a, 310b, 310c** share a common first ion exchange membrane **316** and a common second ion exchange membrane **326**. **FIG. 6** also shows the location of one of the thermally conductive member or fin subassemblies **360**. Fin subassembly **360** includes a thermally conductive plate **362**, a first set of thermally conductive members or fins **366**, which extend toward bicell MEA **310b** and contact cathode **304b**, and a second set of thermally conductive members or fins **364**, which extend away from bicell MEA **310b**. Channels **332a** and **332b** are the fuel flow channels which interconnect the anodes in the multiplexed arrangement **302** shown in **FIG. 6**. Multiplexed arrangement **302** is sealed on both ends by seals **320a, 320b**, preferably formed by the fusing together of first and second ion exchange membranes **316, 326**.

[0066] **FIG. 7** shows a thermally conductive member or fin subassembly **460** which employs a slidable comb **462** for adjusting the configuration of the air conducting channels. The air conducting channels are formed by the spaces between the fins, one of which is designated in **FIG. 7** as fin **466a**. As shown in **FIG. 7**, slidable comb **462** includes a plurality of tines **462a**, which extend into the channels formed by the spaces between the fins.

[0067] **FIG. 8** shows another fin subassembly **560** which employs pivotable baffles (one of which is shown in phantom lines in **FIG. 8** as baffle **574a**). Fin subassembly **560** includes a thermally conductive plate **562**. A plurality of thermally conductive fins **566a, 566b, 566c, 566d** extend from one major surface of plate **562**. In the completed fuel cell assembly incorporating fin subassembly **560**, fins **566a-d** contact the outwardly facing surface of the adjacent cathode (not shown in **FIG. 8**). A plurality of thermally conductive fins **564a, 564b, 564c, 564d, 564e, 564f** extend from the other major surface of plate **562**. Each of fins **564a-f** has a slotted opening formed therein, one of which is shown in **FIG. 8** as slot **570**. A pivotable baffle subassembly, one baffle of which is shown in **FIG. 8** as baffle **574a**, is suspended in the slots by pivot pin **572**. Rotation of baffle **574a** about pivot pin **572** regulates the amount of air flow through the air conducting channels.

[0068] Arrows A in **FIG. 8** show the direction of air flow through the channels formed between fins **566a-d**, and represent the air supply for the electrochemical reaction at the adjacent cathode (not shown). Arrow B in **FIG. 8** shows the direction of air flow through the channels formed between fins **564a-f**, and represents the air supply for conducting heat from the adjacent fuel cell structure (not shown), thereby providing thermal management to the adjacent fuel cell structure.

[0069] **FIG. 9** shows pivotable baffle subassembly **574** for use in conjunction with the fin subassembly **560** in **FIG. 8**. Subassembly **574** includes a plurality of baffles **574a, 574b, 574c** mounted on central pivot pin **572**. **FIG. 10** shows a side view of pivotable baffle subassembly **574**.

[0070] **FIG. 11** shows schematically an electrochemical fuel cell assembly employing ambient air as the oxidant and coolant, which employs external dampers **676, 678** having pivotable baffles **674, 684**, respectively, for adjusting the flow through the air conducting channels **664, 666**. In **FIG. 11**, anode **626**, ion exchange membrane 624 and cathode **616** form the membrane electrode assembly. Fins (not shown) extend from each major surface of plate **662**. The spaces formed between the extending fins form air conducting channels **664, 666**. Dampers **676, 678** include baffles **674, 684**, which are mounted on pivot pins **672, 682**, respectively. Rotation of baffle **674, 684** about the respective pivot pins **672, 682** regulates the amount of air flow through the air conducting channels **664, 666**.

Claims

1. An electrochemical fuel cell assembly (10) comprising:

    a membrane electrode assembly (14) comprising a porous electrically conductive anode (26), a porous electrically conductive cathode (16) having a surface thereof exposed to ambient air, and an ion exchange membrane (24) interposed between said anode (26) and said cathode (16);

    sealant means (50) for forming a gas-impermeable barrier around said anode (26);

    fuel delivery means (44, 46) for supplying a gaseous stream to said anode (26);

    electrical connector means (56a, 66a) for providing an electrical connection to said anode

(26) and to said cathode (16), respectively;

a thermally conductive plate (62a) having oppositely facing first and second major surfaces, said first major surface having a plurality of first thermally conductive members (66a) extending therefrom, said first members (66a) contacting portions of said exposed cathode surface, wherein adjacent ones of said first members (66a) cooperate with said exposed cathode surface to form at least one air conducting channel (78), said second major surface having thermally conductive material (64a) extending therefrom, said thermally conductive material (64a) contacting the atmosphere, whereby heat generated exothermically in said membrane electrode assembly (14) is further dissipated to the atmosphere through said thermally conductive material (64a).

2. The electrochemical fuel cell assembly of claim 1, wherein said plate (62a) is formed as a single planar piece.

3. The electrochemical fuel cell assembly of claim 1, wherein said plate (62a) and said first members (66a) are formed of aluminum, the portions of said first members contacting said cathode surface having an inert metal applied thereto.

4. The electrochemical fuel cell assembly of claim 3, wherein said inert metal is gold.

5. The electrochemical fuel cell assembly of claim 1, wherein said electrical connection means comprises electrical conductors (56a) disposed between said anode (26) and said ion exchange membrane (24), said electrical conductors extending through said sealing means (50).

6. The electrochemical fuel cell assembly of claim 5, wherein said electrical conductors (56a, 56b) are formed from gold wire.

7. The electrochemical fuel cell assembly of claim 1, wherein said thermally conductive material comprises a plurality of thermally conductive members (64a).

8. The electrochemical fuel cell assembly of claim 1, wherein said thermally conductive material comprises a thermally conductive foam.

9. The electrochemical fuel cell assembly of claim 8, wherein said thermally conductive foam is an aluminum foam.

10. The electrochemical fuel cell assembly of claim 1, wherein said fuel delivery means (44, 46) comprises a fuel inlet (46) and a fuel outlet, said fuel outlet directing unreacted components of said gaseous fuel stream away from said anode.

11. The electrochemical fuel cell assembly of claim 1, further comprising a fan for directing said ambient air onto the exposed surface of said porous electrically conductive cathode.

12. The electrochemical fuel cell assembly of claim 1, said assembly further comprising means (78) for accumulating water condensed on said first members (66a).

13. A fuel cell stack (100) comprising:

a plurality of fuel cell assemblies (10a - 10f) as defined in claim 1;

serial connection means (156, 162) for electrically connecting said plurality of fuel cell assemblies in an electrical series having a first assembly (10a) and a last assembly (10f), wherein the anode of each assembly except the last assembly (10f) in said series is electrically connected to the cathode of the next adjacent assembly in said series;

a positive current lead (108) electrically connected to the cathode of said first assembly (10a) in said series; and

a negative current lead (110) electrically connected to the anode of the last assembly (10f) in said series.

14. The fuel cell stack (302) of claim 13, wherein said plurality of fuel cell assemblies (310a - 310c) share a common ion exchange membrane (316).

15. The electrochemical fuel cell assembly of claim 1, wherein said membrane electrode assembly (14) further comprises a second porous electrically conductive cathode (38) having a surface thereof exposed to ambient air and a second ion exchange membrane (34) interposed between said second cathode (38) and said anode (26), said electrical connection means (56a, 56b, 66a, 66b) further providing an electrical connection (66b) to said second cathode (38), said assembly (10) further comprising a second thermally conductive plate (62b) having oppositely facing first and second major surfaces, said first major surface having a plurality of first thermally conductive members (66b) extending therefrom, said first members (66b) contacting portions of said exposed second cathode surface, wherein adjacent ones of said first members (66b) cooper-

ate with said exposed second cathode surface to form at least one air conducting channel (78), said second major surface having thermally conductive material (64b) extending therefrom, said thermally conductive material (64b) contacting the atmosphere, whereby heat generated exothermically in said membrane electrode assembly (14) is further dissipated to the atmosphere through said thermally conductive material (64b).

Patentansprüche

1. Elektrochemische Brennstoffzellenanordnung (10), umfassend:

    eine Membran-Elektroden-Anordnung (14), die eine poröse elektrisch leitende Anode (26), eine poröse elektrisch leitende Kathode (16), deren eine Fläche der Umgebungsluft ausgesetzt ist, und eine zwischen der Anode (26) und der Kathode (16) angeordnete Ionenaustauschmembran (24) umfaßt;

    Dichtmittel (50), um eine gasundurchlässige Barriere um die Anode (26) zu bilden;

    Brennstofflieferenrichtung (44, 46) zum Zuführen eines gasförmigen Stroms zu der Anode (26);

    elektrische Verbindungseinrichtungen (56a, 66a) zur Herstellung einer elektrischen Verbindung zur Anode (26) bzw. zur Kathode (16);

    eine wärmeleitende Platte (62a) mit entgegengesetzter erster und zweiter Hauptfläche, wobei die erste Hauptfläche mehrere sich davon weg erstreckende erste wärmeleitende Elemente (66a) aufweist, diese ersten Elemente (66a) mit Abschnitten der exponierten Kathodenfläche in Kontakt sind, benachbarte dieser ersten Elemente (66a) mit der exponierten Kathodenfläche zusammenwirken, um mindestens einen Luftleitkanal (78) zu bilden, die zweite Hauptfläche sich davon weg erstreckendes wärmeleitendes Material (64a) aufweist, und wobei dieses wärmeleitende Material (64a) mit der Atmosphäre in Kontakt ist, sodaß exotherm in der Membran-Elektroden-Anordnung (14) erzeugte Wärme durch dieses wärmeleitende Material (64a) weiter zur Atmosphäre abgeleitet wird.

2. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, worin die Platte (62a) als ein einziges ebenes Stück ausgebildet ist.

3. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, worin die Platte (62a) und die ersten Elemente (66a) aus Aluminium gebildet sind, wobei auf die mit der Kathodenfläche in Kontakt stehenden Abschnitte der ersten Elemente ein inertes Metall aufgebracht ist.

4. Elektrochemische Brennstoffzellenanordnung nach Anspruch 3, worin das inerte Metall Gold ist.

5. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, worin die elektrische Verbindungseinrichtung zwischen der Anode (26) und der Ionenaustauschmembran (24) angeordnete elektrische Leiter (56a) umfaßt, wobei sich diese elektrischen Leiter durch das Dichtmittel (50) erstrecken.

6. Elektrochemische Brennstoffzellenanordnung nach Anspruch 5, worin die elektrischen Leiter (56a, 56b) aus Golddraht gebildet sind.

7. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, worin das wärmeleitende Material mehrere wärmeleitende Elemente (64a) umfaßt.

8. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, worin das wärmeleitende Material aus einem wärmeleitenden Schaum besteht.

9. Elektrochemische Brennstoffzellenanordnung nach Anspruch 8, worin der wärmeleitende Schaum ein Aluminiumschaum ist.

10. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, worin die Brennstofflieferenrichtung (44, 46) einen Brennstoffeinlaß (46) und einen Brennstoffauslaß aufweist, wobei der Brennstoffauslaß nicht umgesetzte Komponenten des gasförmigen Brennstoffstroms von der Anode wegleitet.

11. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, die weiter ein Gebläse aufweist, das Umgebungsluft auf die exponierte Fläche der porösen elektrisch leitenden Kathode richtet.

12. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, die weiter Einrichtungen (78) zum Sammeln von auf den ersten Elementen (66a) kondensiertem Wasser aufweist.

13. Brennstoffzellenstapel (100) umfassend:

    eine Vielzahl von Brennstoffzellenanordnungen (10a-10f), wie im Anspruch 1 definiert;

    Serienverbindungseinrichtungen (156, 162) zur elektrischen Verbindung dieser Vielzahl von Brennstoffzellenanordnungen in elektri-

scher Serienschaltung, die eine erste Anordnung (10a) und eine letzte Anordnung (10f) aufweist, wobei die Anode jeder Anordnung mit Ausnahme der letzten (10f) in dieser Serienschaltung elektrisch mit der Kathode der nächsten benachbarten Anordnung in der Serienschaltung verbunden ist;

eine positive Stromleitung (108), die elektrisch mit der Kathode der ersten Anordnung (10a) in der Serienschaltung verbunden ist; und

eine negative Stromleitung (110), die elektrisch mit der Anode der letzten Anordnung (10f) in der Serienschaltung verbunden ist.

14. Brennstoffzellenstapel (302) nach Anspruch 13, worin die Vielzahl von Brennstoffzellenanordnungen (310a - 310c) eine gemeinsame Ionenaustauschmembran (316) teilt.

15. Elektrochemische Brennstoffzellenanordnung nach Anspruch 1, worin die Membran-Elektroden-Anordnung (14) weiter eine zweite poröse elektrisch leitende Kathode (38), deren eine Fläche der Umgebungsluft ausgesetzt ist, und eine zweite, zwischen dieser zweiten Kathode (38) und der Anode (26) angeordnete Ionenaustauschmembran (34) aufweist, die elektrischen Verbindungseinrichtungen (56a, 56b, 66a, 66b) weiter eine elektrische Verbindung (66b) zur zweiten Kathode (38) herstellen, die Anordnung (10) weiter eine zweite wärmeleitende Platte (62b) mit entgegengesetzter erster und zweiter Hauptfläche aufweist, wobei die erste Hauptfläche mehrere sich davon weg erstreckende erste wärmeleitende Elemente (66b) aufweist, diese ersten Elemente (66b) mit Abschnitten der exponierten zweiten Kathodenfläche in Kontakt sind, benachbarte dieser ersten Elemente (66b) mit der exponierten zweiten Kathodenfläche zusammenwirken, um mindestens einen Luftleitkanal (78) zu bilden, die zweite Hauptfläche sich davon weg erstreckendes wärmeleitendes Material (64b) aufweist, und wobei dieses wärmeleitende Material (64b) mit der Atmosphäre in Kontakt ist, sodaß exotherm in der Membran-Elektroden-Anordnung (14) erzeugte Wärme durch dieses wärmeleitende Material (64b) weiter zur Atmosphäre abgeleitet wird.

Revendications

1. Assemblage de cellule électrochimique à combustible (10) comprenant :

un assemblage d'électrodes à membrane (14) comprenant une anode poreuse électriquement conductrice (26), une cathode poreuse électriquement conductrice (16) ayant une surface de celle-ci exposée à l'air ambiant et une membrane échangeuse d'ions (24) intercalée entre ladite anode (26) et ladite cathode (16);
un moyen d'étanchéité (50) pour former une barrière imperméable au gaz autour de ladite anode (26);
un moyen d'alimentation en combustible (44, 46) pour fournir un courant gazeux à ladite anode (26);
un moyen de connecteur électrique (56a, 66a) pour foumir respectivement une connexion électrique à ladite anode(26) et à ladite cathode (16);
une plaque thermiquement conductrice (62a) présentant des première et seconde surfaces principales se faisant face, ladite première surface principale présentant de nombreux premiers éléments thermiquement conducteurs (66a) s'étendant à partir de celle-ci, lesdits premiers éléments (66a) étant en contact avec des parties de ladite surface de cathode exposée, dans lequel les éléments adjacents aux dits premiers éléments (66a) coopèrent avec ladite surface de cathode exposée pour former au moins un canal conduisant l'air (78), ladite seconde surface principale présentant un matériau thermiquement conducteur (64a) s'étendant à partir de celle-ci, ledit matériau thermiquement conducteur (64a) étant en contact avec l'atmosphère, la chaleur exothermiquement produite dans ledit assemblage d'électrodes à membrane (14) est par là en outre dissipée vers l'atmosphère par l'intermédiaire dudit matériau thermiquement conducteur (64a).

2. Assemblage de cellule électrochimique à combustible selon la revendication 1, dans lequel ladite plaque (62a) est formée comme une seule pièce plane.

3. Assemblage de cellule électrochimique à combustible selon la revendication 1, dans lequel ladite plaque (62a) et lesdits premiers éléments (66a) sont constitués d'aluminium, les parties desdits premiers éléments étant en contact avec ladite surface de cathode présentant un métal inerte appliqué à celles-ci.

4. Assemblage de cellule électrochimique à combustible selon la revendication 3, dans lequel ledit métal inerte est l'or.

5. Assemblage de cellule électrochimique à combustible selon la revendication 1, dans lequel ledit moyen de connexion électrique comprend des conducteurs électriques (56a) disposés entre ladite anode (26) et ladite membrane échangeuse d'ions (24), lesdits conducteurs électriques s'étendant à

travers ledit moyen d'étanchéité (50).

6. Assemblage de cellule électrochimique à combustible selon la revendication 5, dans lequel lesdits conducteurs électriques (56a, 56b) sont constitués par un câble en or.

7. Assemblage de cellule électrochimique à combustible selon la revendication 1, dans lequel ledit matériau thermiquement conducteur comprend de nombreux éléments thermiquement conducteurs (64a).

8. Assemblage de cellule électrochimique à combustible selon la revendication 1, dans lequel ledit matériau thermiquement conducteur comprend une mousse thermiquement conductrice.

9. Assemblage de cellule électrochimique à combustible selon la revendication 8, dans lequel ladite mousse thermiquement conductrice est une mousse d'aluminium.

10. Assemblage de cellule électrochimique à combustible selon la revendication 1, dans lequel ledit moyen d'alimentation en combustible (44, 46) comprend une entrée de combustible (46) et une sortie de combustible, ladite sortie de combustible évacuant des constituants n'ayant pas réagi dudit courant de combustible gazeux de ladite anode.

11. Assemblage de cellule électrochimique à combustible selon la revendication 1, comprenant en outre une soufflante pour diriger ledit air ambiant sur la surface exposée de ladite cathode poreuse électriquement conductrice.

12. Assemblage de cellule électrochimique à combustible selon la revendication 1, ledit assemblage comprenant en outre un moyen (78) pour accumuler l'eau condensée sur lesdits premiers éléments (66a).

13. Empilement de cellules à combustible (100) comprenant :

    de nombreux assemblages de cellules à combustible (10a-10f) selon la revendication 1 ;
    un moyen de connexion en série (156, 162) pour raccorder électriquement lesdits nombreux assemblages de cellules à combustible en une série électrique présentant un premier assemblage (10a) et un dernier assemblage (10f), dans lequel l'anode de chaque assemblage à l'exception du dernier assemblage (10f) dans ladite série est électriquement raccordée à la cathode de l'assemblage adjacent suivant dans ladite série ;
    une ligne d'alimentation de courant positive (108) électriquement raccordée à la cathode dudit premier assemblage (10a) dans ladite série ; et
    une ligne d'alimentation de courant négative (110) électriquement raccordée à l'anode du dernier assemblage (10f) dans ladite série.

14. Empilement de cellules à combustible (302) selon la revendication 13, dans lequel lesdits nombreux assemblages de cellules à combustible (310a-310c) partagent une membrane échangeuse d'ions commune (3 16).

15. Assemblage de cellule électrochimique à combustible selon la revendication 1, dans lequel ledit assemblage d'électrodes à membrane (14) comprend en outre une seconde cathode poreuse électriquement conductrice (38) présentant une surface de celle-ci exposée à l'air ambiant et une seconde membrane échangeuse d'ions (34) intercalée entre ladite seconde cathode (38) et ladite anode (26), ledit moyen de connexion électrique (56a, 56b, 66a, 66b) fournissant en outre une connexion électrique (66b) à ladite seconde cathode (38), ledit assemblage (10) comprenant en outre une seconde plaque thermiquement conductrice (62b) présentant des première et seconde surfaces principales se faisant face, ladite première surface principale présentant de nombreux premiers éléments thermiquement conducteurs (66b) s'étendant à partir de celle-ci, lesdits premiers éléments (66b) étant en contact avec des parties de ladite surface de la seconde cathode exposée, dans lequel les éléments adjacents desdits premiers éléments (66b) coopèrent avec ladite surface de la seconde cathode exposée pour former au moins un canal conduisant l'air (78), ladite seconde surface principale présentant un matériau thermiquement conducteur (64b) s'étendant à partir de celle-ci, ledit matériau thermiquement conducteur (64b) étant en contact avec l'atmosphère, la chaleur exothermiquement produite dans ledit assemblage d'électrodes à membrane (14) est par là en outre dissipée vers l'atmosphère par l'intermédiaire dudit matériau thermiquement conducteur (64b).

Fig. 1

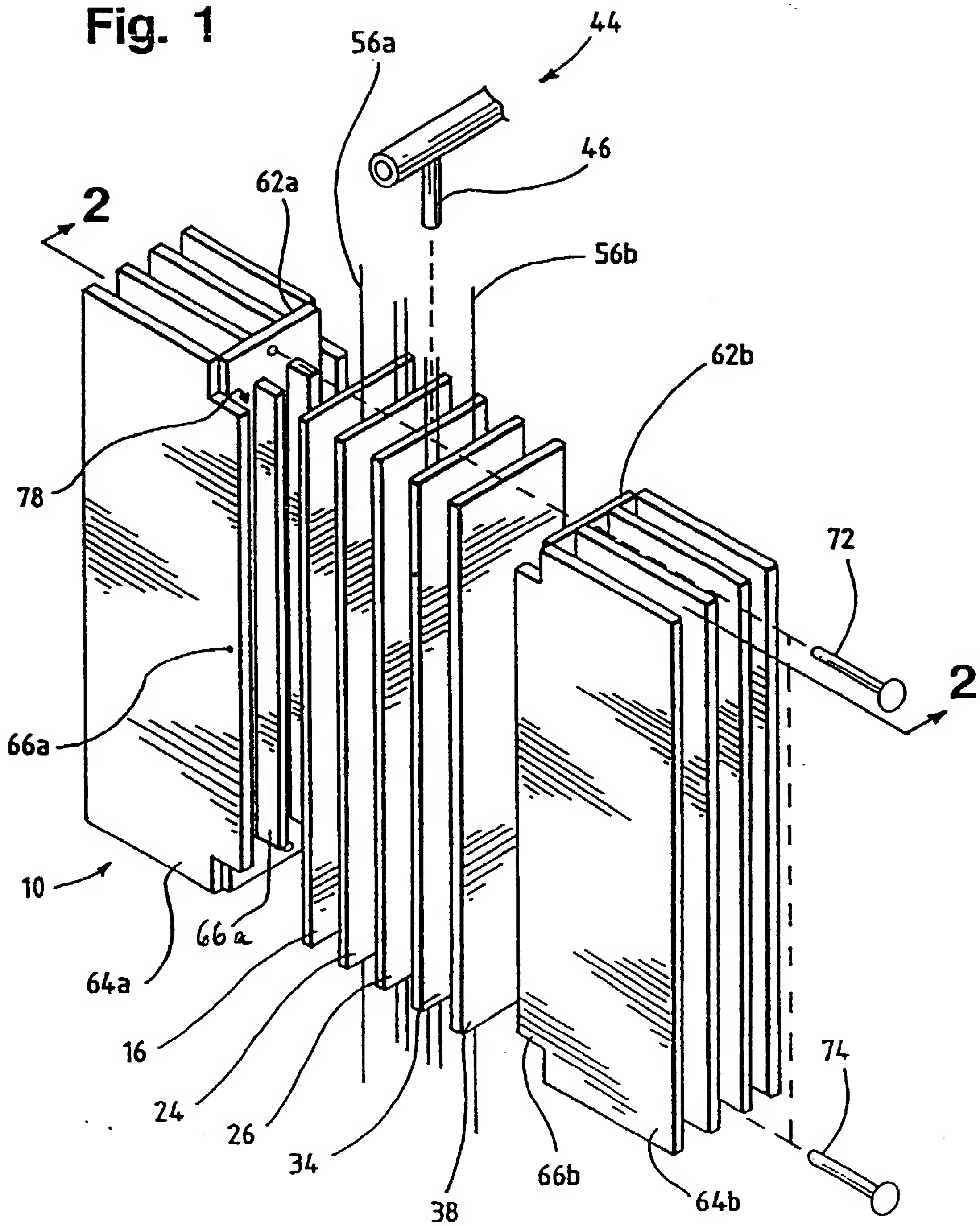

44
56a
46
62a
2
56b
62b
78
72
2
66a
10
66a
64a
16
24
26
34
38
66b
64b
74

Fig. 2A

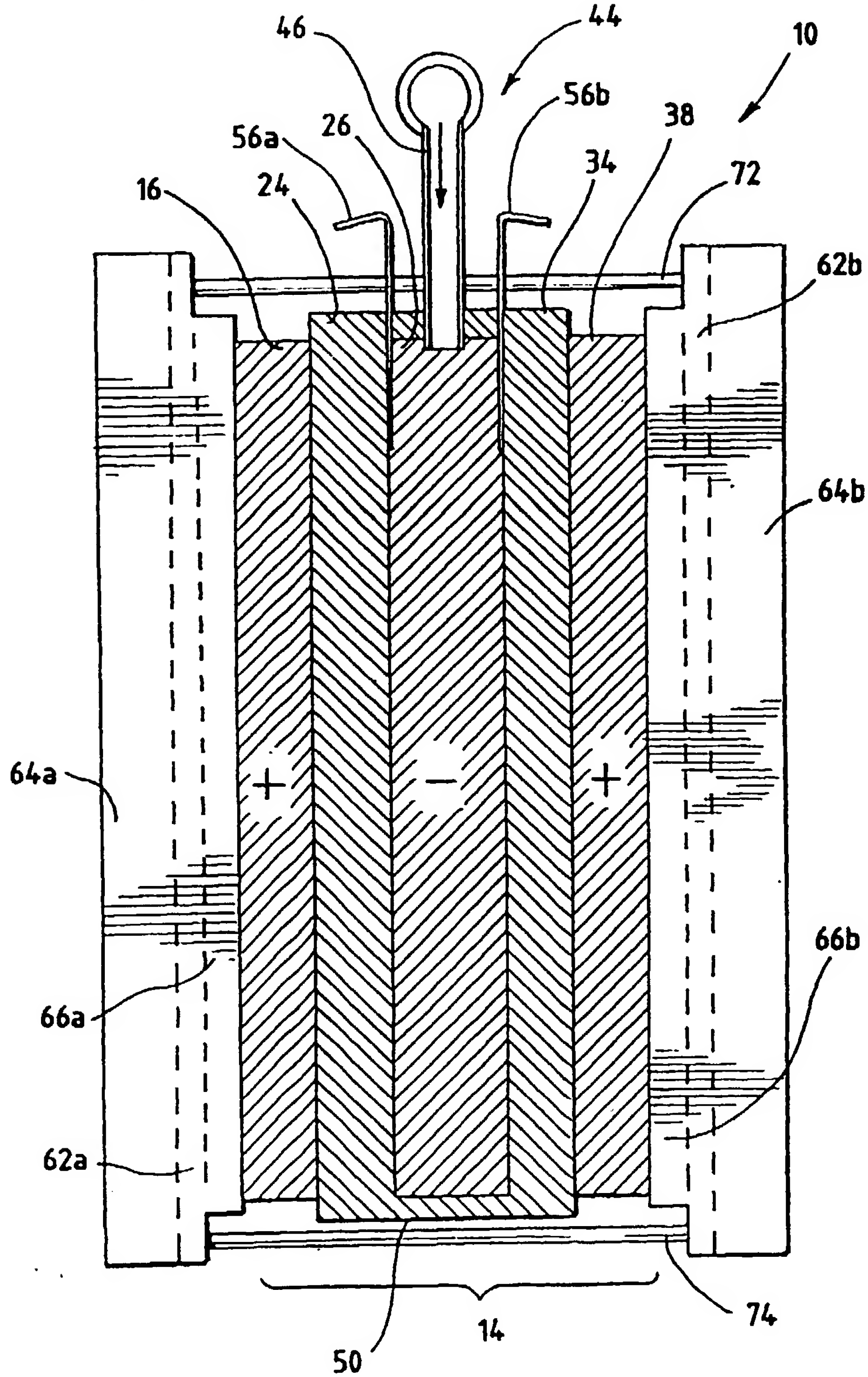


Fig. 2B

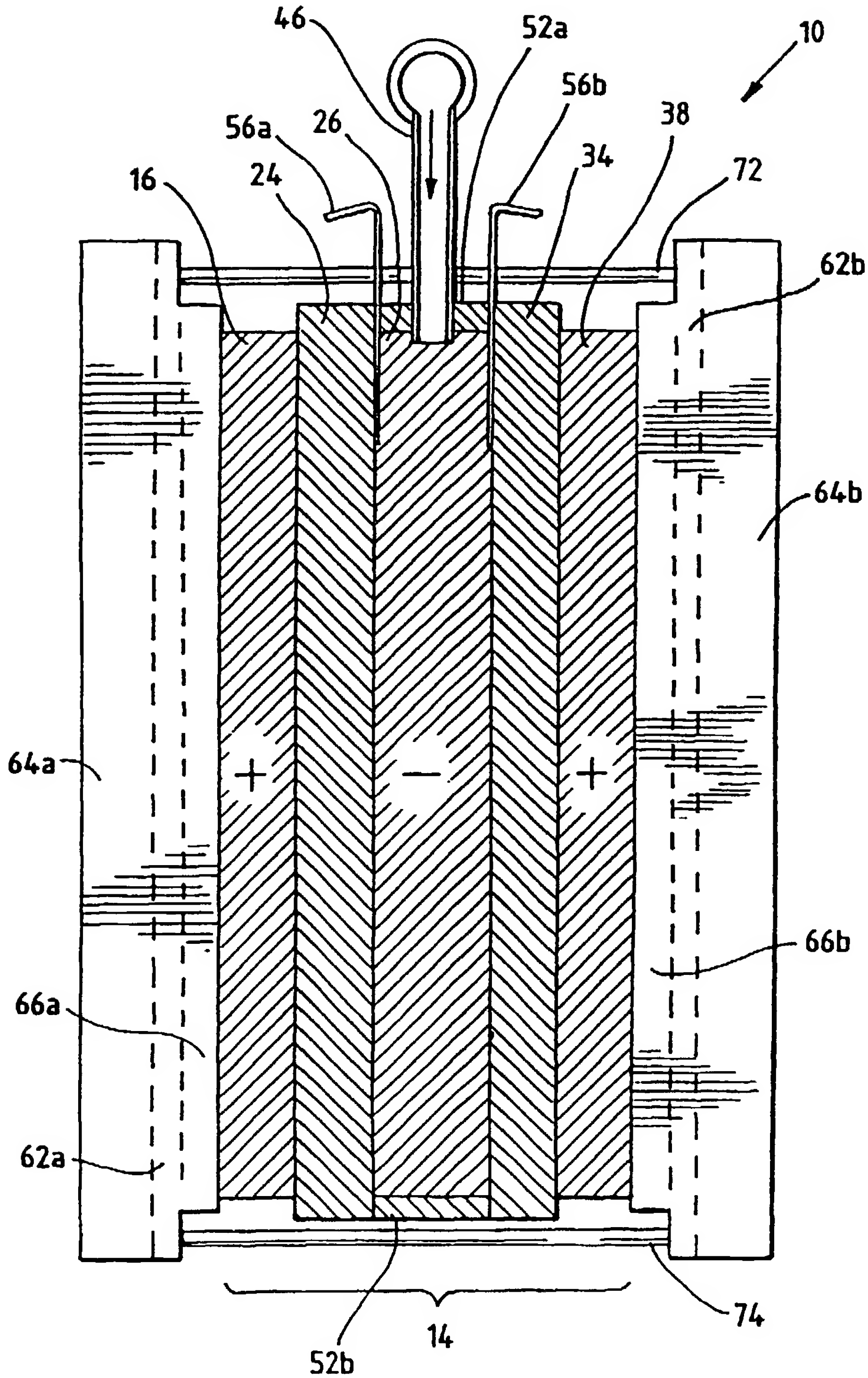
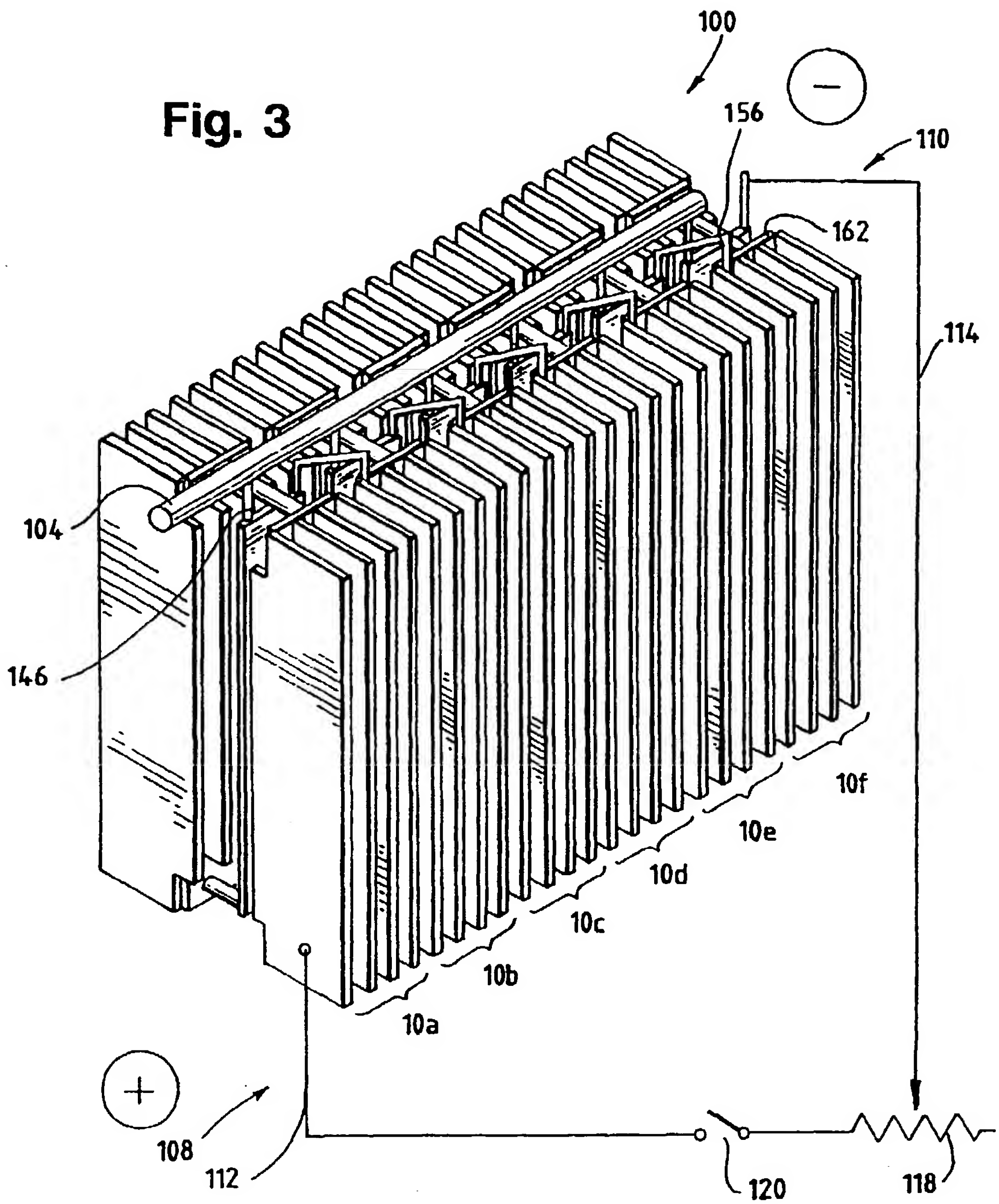

46
52a
10
56b
56a
26
38
34
16
24
72
62b
64b
64a
+
−
+
66b
66a
62a
74
14
52b

Fig. 3


100
156
110
162
114
104
146
10f
10e
10d
10c
10b
10a
108
112
120
118

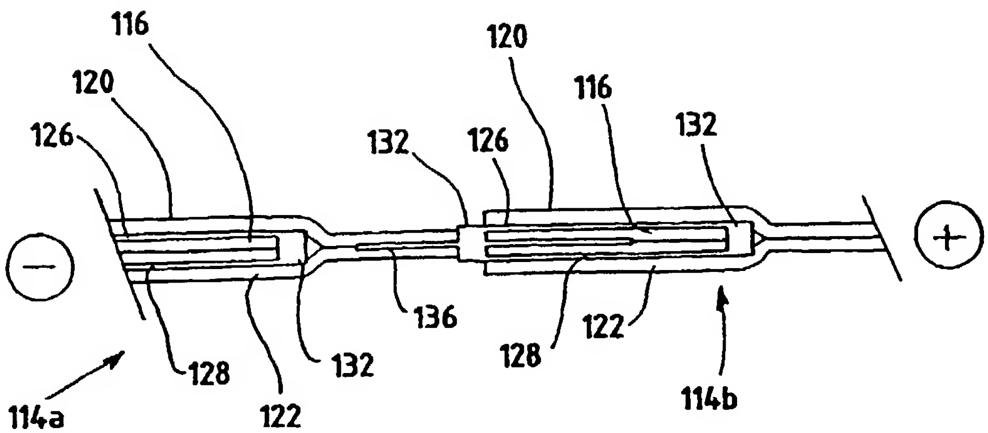

116
120
126
120
116
132
126
132
136
128
122
132
128
122
114a
122
114b

Fig. 4A

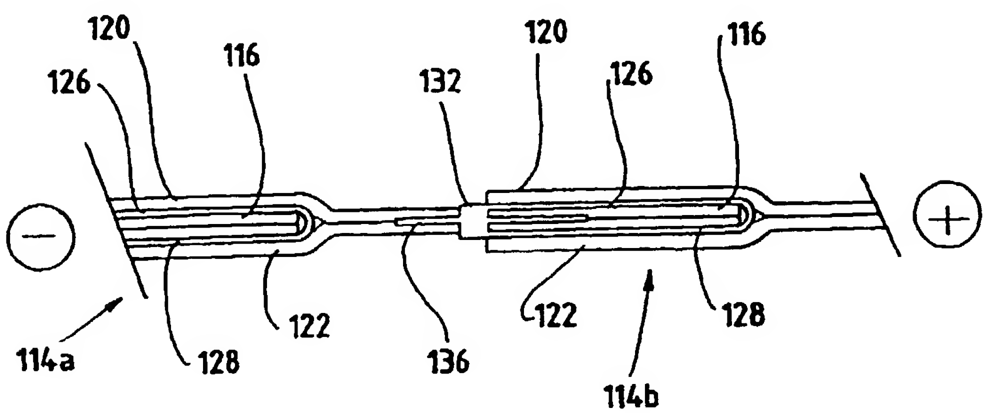
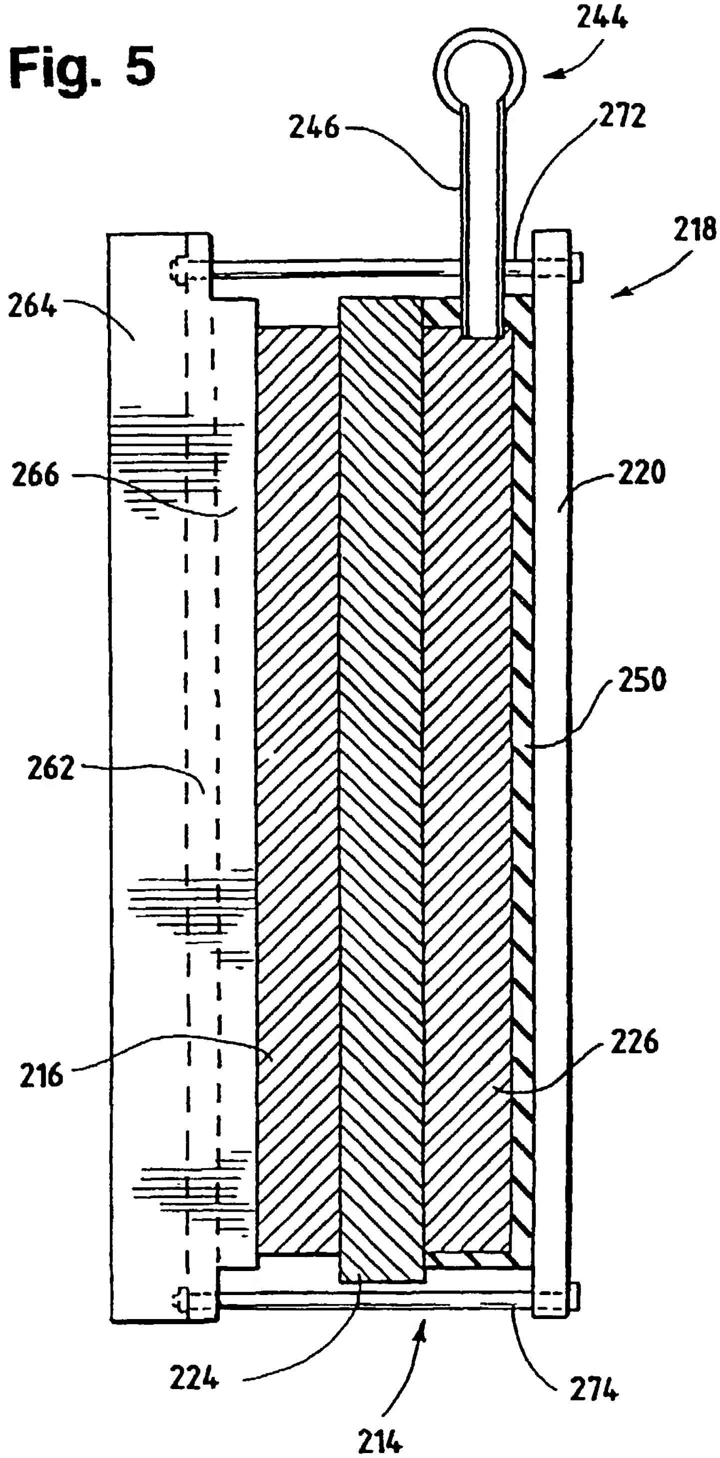

120
126
116
120
116
126
132
122
128
122
128
136
114a
114b

Fig. 4B

Fig. 5



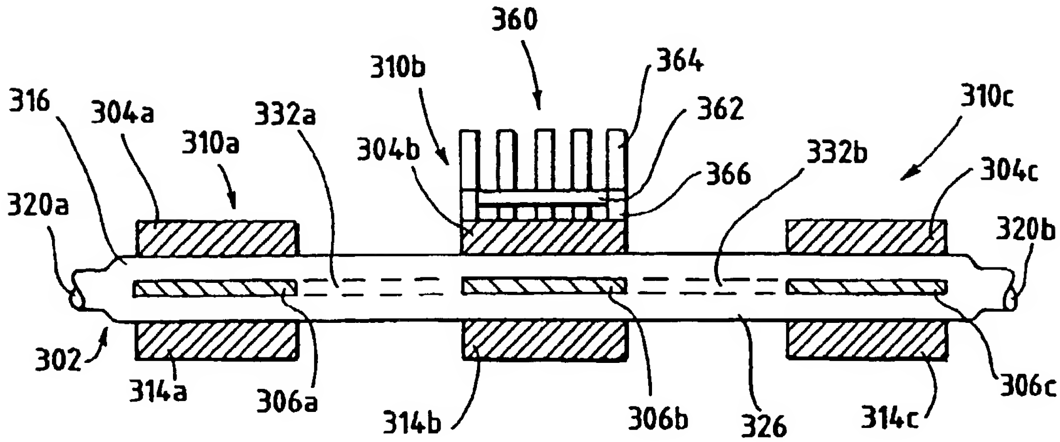

360
310b
364
316
304a
332a
362
310c
310a
304b
366
332b
304c
320a
320b
302
314a
306a
314b
306b
326
314c
306c

Fig. 6

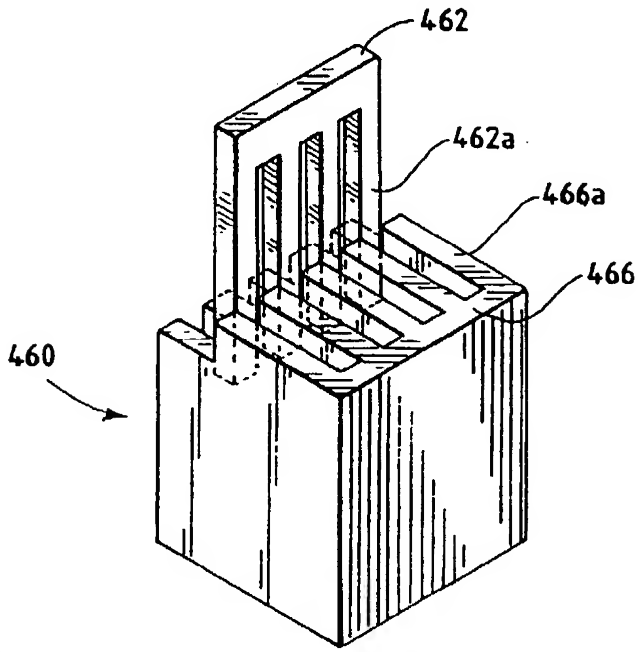

462
462a
466a
466
460

Fig. 7

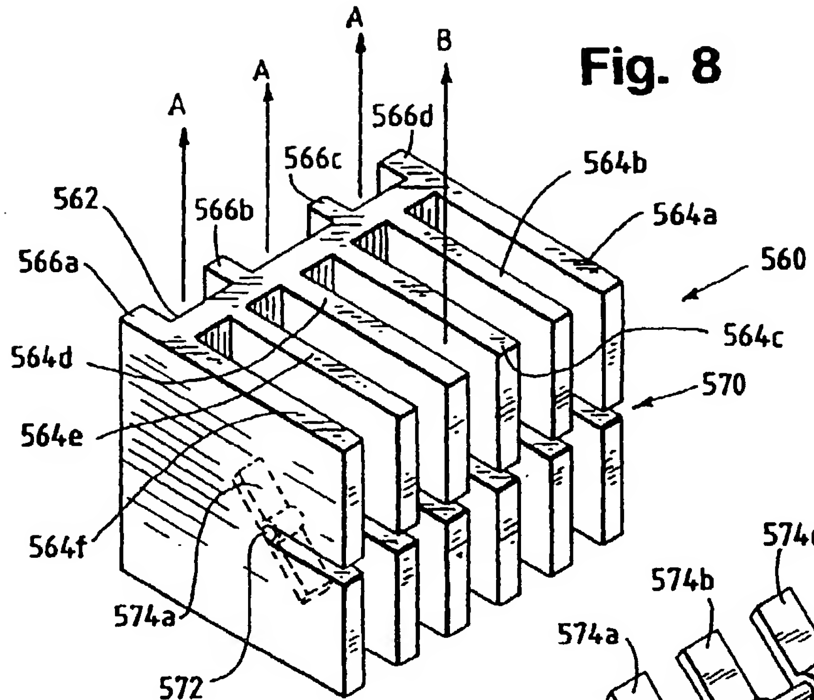


Fig. 8

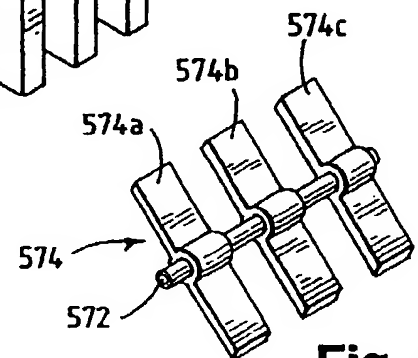


Fig. 9

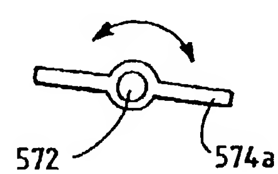


Fig. 10

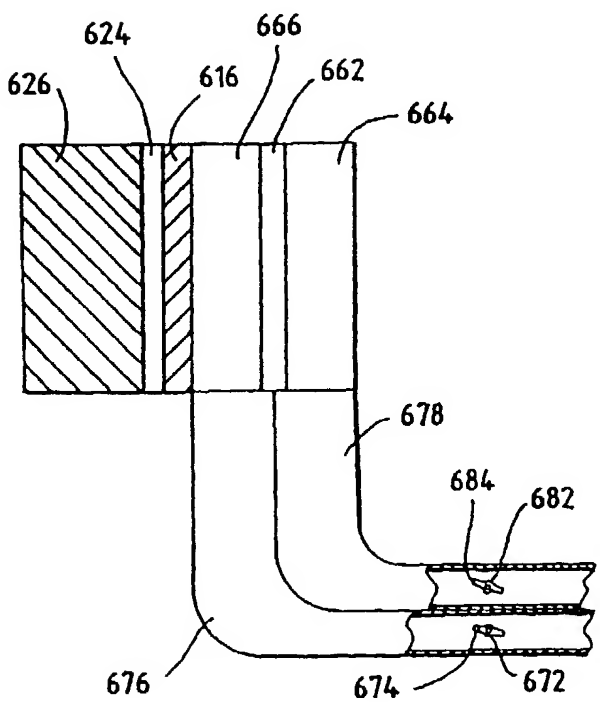


Fig. 11